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THRESHOLD TEAR STRENGTH OF ELASTOMERS

by

A. N. Gent and R. H. Tobias

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) Tear strengths have been measured for a wide variety of molecular networks under threshold conditions; i.e., at high temperatures, low rates of tearing, and with swollen samples. For all of the polymers examined, the threshold tear strength was found to be proportional to the square root of the average molecular weight M_c of network strands, in agreement with theory. However, for the same M_c and hence for similar values of elastic		

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modulus, different polymers showed major differences in threshold tear strength. The tear strength of polydimethylsiloxane networks was only about one-third as large as that for networks of polybutadiene and cis-polyisoprene and the values obtained for polyphosphazene networks were only about one-fifth as large, at the same M_c . These striking differences are attributed to differences in network strand length and extensibility for the same molecular weight. The threshold tear strengths are shown to be in satisfactory quantitative agreement with theoretically-predicted values on this basis.

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THRESHOLD TEAR STRENGTH OF ELASTOMERS

by

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Introduction

The tear strength of an elastomeric material has been shown to reach a lower limit, termed here the threshold strength, when dissipative processes are minimized (1,2). The threshold value can be determined experimentally at low rates of tearing, at high temperatures, and when the material is highly swollen with a low-viscosity liquid. Under these near equilibrium conditions, experimental tear strengths are found to reach minimum values of 40-80 J/m² (1,2). Lake and Thomas (3) have developed a simple theoretical treatment to predict the magnitude of the threshold tear strength for elastomers from the length of the molecular strands comprising a network and the dissociation energy of the chemical bonds comprising each strand. Expressed as the energy T_o required to tear through a unit area of the material, their theoretical result is

$$T_o = KM_c^{1/2} \quad (1)$$

where M_c is the mean molecular weight of the network

strands and \underline{K} is a constant involving the effective mass, length and flexibility of a single main-chain bond, the density of the polymer and the dissociation energy of the weakest bond. For C-C molecular strands \underline{K} is predicted to be about $0.3 \text{ J/m}^2/(\text{molecular weight unit})^{1/2}$. Experimental values of \underline{T}_0 for randomly crosslinked networks of polybutadiene were found to be consistent with equation 1 when \underline{K} was given a somewhat higher value, about $1.0 \text{ J/m}^2/(\text{molecular weight unit})^{1/2}$. Apart from this numerical discrepancy, the threshold strength of polybutadiene networks seems to be reasonably well accounted for (2).

Measurements have now been carried out on a number of elastomers, of widely differing chemical constitution. They are: cis-polyisoprene (cis-PI), trans-polyisoprene (trans-PI), polydimethylsiloxane (PDMS) and a fluoroalkoxy-substituted polyphosphazene (PNF). In each case, networks were made of a wide range of strand lengths, by a random crosslinking process, and the threshold tear strengths determined by careful measurements at high temperatures and low rates of tearing. Tear strengths were also measured for samples swollen highly with low-viscosity fluids, for comparison with the results obtained with unswollen materials. The results are given in the following sections of this paper and compared with the predictions of the Lake-Thomas theory.

A brief outline of the Lake-Thomas theory is now given, in order to draw attention to the main molecular parameters which appear in the coefficient K relating the threshold tear strength to \underline{M}_c in equation 1. It is assumed that a number \underline{N}' of network strands pass through a randomly-chosen fracture plane of unit area and that this number must be broken for the fracture to propagate. The work required to break them is \underline{T}_0 . Each strand is regarded as of uniform length, composed of \underline{n} main-chain atoms, each with an associated average molecular weight of \underline{M}_0 . Thus, the molecular weight \underline{M}_c of a strand is given by \underline{nM}_0 , and its dissociation energy by \underline{nU} where \underline{U} is the dissociation energy of a main-chain bond. Hence,

$$\underline{T}_0 = \underline{N}' \underline{M}_0 \underline{U} \quad (2)$$

In order to determine the number \underline{N}' of strands crossing the fracture plane in terms of the number \underline{N} of network strands per unit volume, it is assumed that only those strands lying within a volume element defined by the fracture plane itself and a perpendicular distance \underline{R} , equal to the r.m.s. distance between strand ends in the undeformed state, need to be considered. Furthermore, only about one-third of these strands will actually cross the fracture plane. The other two-thirds will lie generally parallel to the plane and hence escape fracture. Thus,

$$\underline{N}' (= \underline{N} \underline{R} / 3) = 1/3 (\rho \underline{A} / \underline{M}_c) \underline{R}$$

where ρ is the density of the polymer and \underline{A} is Avogadro's number.

A more precise computation of \underline{N}' yields a numerical factor of $(3/8)^{1/2}$ in place of $1/3$.

The distance \underline{R} is given by $\underline{n}_r^{1/2} \underline{l}_r$, where \underline{n}_r is the number of links in a hypothetical chain of freely-jointed links chosen to have the same value of \underline{R} and fully-stretched length \underline{l} as the "real" chain of \underline{n} main-chain bonds, each of projected length \underline{l} (4). Typically, the length \underline{l}_r of an equivalent freely-jointed link is several main-chain bonds and, correspondingly, the number \underline{n}_r of random links in the equivalent chain is several times smaller than the number \underline{n} of bonds. The parameter $q = \underline{l}_r / \underline{l} = \underline{n} / \underline{n}_r$ is a measure of chain stiffness. Estimates of q can be obtained from measurements of the stress-optical coefficient (4); they range from 5-10 main-chain bonds for various elastomer polymers.

On substituting in equation 2 for \underline{N}' , \underline{T}_0 is finally obtained as

$$\underline{T}_0 = (3/8)^{1/2} \rho A U q^{1/2} \underline{l} M_c^{1/2} / M_0^{3/2}.$$

The coefficient \underline{K} in equation 1 is thus given by

$$\underline{K} = (3/8)^{1/2} \rho A U q^{1/2} \underline{l} / M_0^{3/2}. \quad (3)$$

Values of \underline{K} calculated from equation 3 are compared with experimentally-determined values in the final section of this paper.

2. Experimental details

(a) Materials

(1) Cis-polyisoprene (cis-PI) and trans-polyisoprene (trans-PI)

Samples of 96% cis-1,4 - polyisoprene (Natsyn 2200, Goodyear Tire and Rubber Company) and trans 1,4 - polyisoprene (Trans-Pip, Polysar Inc.) were mixed with various amounts of dicumyl peroxide (DiCup R, Hercules Chemical Company). They were then pressed into sheets, about 1.5 mm thick, and cross-linked by heating them for 2 hr at 150°C. Similar samples were also prepared from 100% cis 1,4 - polyisoprene (natural rubber, SMR-5L) but, as described later, it was not found possible to determine the threshold tear strength for these samples with comparable precision when the degree of cross-linking was low.

(ii) Polybutadienes

Samples of cis 1,4 - polybutadiene (Cis - 4, Phillips Petroleum Company) and a cis: trans: vinyl copolymer (36:54:10, Diene 35 NFA, Firestone Tire and Rubber Company) have been examined previously (2). The results are included here for comparison with those obtained for other elastomeric materials.

(iii) Polydimethylsiloxane (PDMS)

This polymer was supplied by General Electric Company. The number-average molecular weight \bar{M}_n was 430,000 g/g-mole.

It was also mixed with various amounts of dicumyl peroxide and crosslinked by heating for 2 h at 150°C.

(iv) Fluoroalkoxy-substituted polyphosphazene (PNF)

Polyphosphazene (Phosphonitrilic Fluoroelastomer PNF-200, Firestone Tire and Rubber Company) was mixed with various amounts of dicumyl peroxide (DiCup R, Hercules Chemical Company) and crosslinked by heating for 2 h at 150°C.

(i) Measurement of network strand molecular weight $\underline{M_c}$

Values of the Mooney-Rivlin elastic coefficients $\underline{C_1}$ and $\underline{C_2}$ were determined from stress-strain relations in tension (4), determined at ambient temperature. The values obtained are given in Table 1, together with corresponding values of the small-strain elastic modulus (Young's modulus) $\underline{E} = 6 (\underline{C_1} + \underline{C_2})$. The extensibility of the PNF materials was too small to permit an accurate determination of $\underline{C_1}$ and $\underline{C_2}$. Values of \underline{E} were obtained in these cases from the initial slopes of the stress-strain relations.

According to the statistical theory of rubberlike elasticity, \underline{E} is directly related to the average network strand molecular weight $\underline{M_c}$ (4),

$$\underline{E} = 3 \underline{\rho} RT / \underline{M_c} \quad (4)$$

where $\underline{\rho}$ is the density of the elastomer, \underline{R} is the gas constant and \underline{T} is absolute temperature. Values of $\underline{M_c}$ calculated by means of equation 4 are given in Table 1.

The exact relationship between the chemical structure of the network and the elastically-effective strand population is still subject to debate. It has been assumed here that

those network strands that govern the small-strain elastic behavior are also responsible for the tear strength under threshold conditions, so that values of \underline{M}_c calculated from equation 4 are appropriately employed in equation 1. This assumption ignores the non-Gaussian behavior of rubber materials. It has been claimed that the \underline{C}_1 term is directly proportional to the density of network strands and that the non-Gaussian \underline{C}_2 term arises from constraints on their elastic response which become less important at high strains. From this point of view, it would be more appropriate to calculate \underline{M}_c from \underline{C}_1 ,

$$\underline{C}_1 = \rho RT / 2 \underline{M}_c$$

for comparison with tear strengths, which are inevitably associated with high strains. Had the elastic coefficient \underline{C}_1 been used instead of \underline{E} for calculating \underline{M}_c the values obtained would have been generally about twice as large and values of the constant \underline{K} from equation 1 would then have been about 30% lower than those discussed below. The general form of the results and the relative rankings of the different elastomers would not have been altered, however.

(vi) Measurement of threshold tear strength

Rectangular strips, about 60 mm long, 10 mm wide and 1.4 mm thick were scored along a central line to a depth of about 0.7 mm, leaving about one-half of the original thickness to be torn through. Tearing was generally found to take place at an angle of approximately 45° to the sheet

thickness, as shown schematically in Figure 1. The tear energy T was calculated from the measured tear force F by the relation (1, 2)

$$T = 2 \lambda_s^2 F/t \quad (5)$$

where λ_s is the linear swelling ratio of the sample and t is the width of the tear path, measured on the torn strip after tearing was completed. The term λ_s^2 in equation 5 accounts for the reduced number of network strands crossing the tear plane in a swollen specimen. For unswollen samples, $\lambda_s = 1$.

The swelling liquids used were m-xylene or paraffin oil with PI and PB networks, m-xylene or silicone oil with PDMS networks, and dibutyl sebacate with PNF networks. Samples were torn while immersed in a water bath, at temperatures between 70°C and 90°C. The water effectively prevented evaporation of the swelling liquid during tearing.

For natural rubber samples swollen with paraffin oil it was found necessary to use much higher test temperatures, in the range 90°C - 180°C, in order to approach a lower limit in tear strength. For the lightly-crosslinked materials the tear strength did not reach a lower limit even at temperatures of 150°C, Figure 2, and at temperatures much above this, rapid deterioration occurred. It is thought that strain-induced crystallization was present, even at high temperatures and in the swollen state, strengthening these materials in comparison with wholly-amorphous elastomers.

Swollen samples of cis-PI containing about 96% cis units were found to reach well-defined threshold values at approximately 140°C. Presumably the somewhat smaller cis content reduced the tendency to crystallize on stretching, so that these materials were completely amorphous during tearing at 140°C and above.

Good agreement was obtained between values of \underline{T} determined with swollen and unswollen samples, provided that sufficiently high test temperatures were used. Some representative results for PDMS materials are given in Table 2. Mean values of \underline{T} for swollen and unswollen materials have been taken as measures of the threshold tear strength \underline{T}_0 . They are given in Table 1 for all of the materials examined.

3. Experimental results and discussion

Experimentally-determined values of the threshold tear strength T_0 are plotted against the elastic modulus E in Figure 3 and against corresponding values of M_c in Figure 4, using logarithmic scales for both axes. The results are in reasonably good agreement with linear relations in all cases, with slopes of $-1/2$ when plotted against E and $+1/2$ when plotted against M_c . Thus, the general form of the results is in good agreement with the theory of Lake and Thomas (3).

It is noteworthy that at similar values of E , the threshold tear strength of the hydrocarbon elastomers are all rather similar in magnitude but they are much larger than for PDMS and PNF, by a factor of about three. Marked differences are also shown at similar values of M_c , Figure 4, by a factor of about three for PDMS and about five for PNF. These differences are attributed to differences in the molecular constants which govern the coefficient K relating the threshold tear strength to M_c . Estimated values of the various molecular constants are listed in Table 3, together with the values of K calculated from them by means of equation 3. Experimentally-determined values of K , taken from the linear relations for each elastomer shown in Figure 4, are included in Table 3 for comparison with theoretically-derived results.

The agreement is reasonably good, both in absolute magnitude and in the relative ranking of the various elastomers.

As there are no fitting constants in the theory, this agreement must be regarded as quite satisfactory and indicates that the main molecular parameters governing the tear strength of elastomers under threshold conditions have been taken into account.

The large effect of the mass per main-chain atom is particularly noteworthy. It appears to be the principal factor responsible for the striking differences between the tear strengths of the hydrocarbon elastomers, PI and PB, and those of the inorganic elastomers, PDMS and PNF.

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Clarendon Press, Oxford, 1958.

Table 1. Threshold tear strength T_o of molecular networks
with varying M_c

Dicumyl peroxide (%)	C_1 (kPa)	C_2 (kPa)	E (kPa)	$M_c \times 10^{-3}^{(a)}$ (g/g-mole)	T_o (J/m ²)
		<u>NR</u>			
1	95	73	1010	6.7	<u>ca</u> 150
2	182	63	1470	4.6	<u>ca</u> 90
3	254	83	2020	3.3	62 ± 7
4	337	68	2430	2.8	52 ± 5
5	424	122	3275	2.1	43 ± 3
		<u>cis-PI</u>			
1	121	71	1150	5.8	108 ± 9
2	197	78	1650	4.1	63 ± 4
4	387	48	2610	2.6	51 ± 5
		<u>trans-PI</u>			
2	155	132	1720	4.8	60 ± 5
3	228	91	1915	4.3	48 ± 4
4	384	63	2680	3.1	38 ± 5
		<u>cis-PB</u> (b)			
0.5	182	174	2135	3.1	81 ± 8
2.0	455	160	3690	1.8	58 ± 8
		<u>PB</u> (b)			
0.025	44	148	1150	5.8	78 ± 8
0.05	110	148	1550	4.3	71 ± 9
0.2	332	148	2880	2.3	45 ± 5
0.4	570	145	4290	1.6	40 ± 6
		<u>PDMS</u>			
1.0	9	20	175	41.0	78 ± 6
1.2	14	23	220	32.2	74 ± 3
1.5	19	29	290	24.8	62 ± 3
1.75	21	30	305	23.3	56 ± 3
2.0	26	31	340	20.9	48 ± 3
2.5	31	31	370	19.2	46 ± 2
2.75	32	35	400	17.8	44 ± 3
3.0	36	33	415	17.2	42 ± 3
4.0	45	25	420	17.0	39 ± 2
		<u>PNF</u>			
0.5			225	56	55 ± 8
1.0			340	37	45 ± 5
1.5			365	34	41 ± 5
2.0			395	32	40 ± 4
3.2 (c)			435	29	38 ± 4

^aCalculated from E using equation 4.

^bTaken from reference 2.

^c2% Vul-Cup R (Hercules, Inc.), equivalent to 3.2% dicumyl peroxide.

Table 2. Effect of swelling with m-xylene on the threshold tear strength of PDMS networks

Dicumyl peroxide %	T_0 (Unswollen) (J/m ²)	λ_s	T_0 (Swollen) (J/m ²)	$\lambda_s^2 T_0$ (Swollen) (J/m ²)
1.0	89 \pm 8	2.22	17.2 \pm 1.7	85 \pm 10
1.2	79 \pm 5	1.94	19.8 \pm 2.0	75 \pm 8
1.5	62 \pm 5	1.91	17.3 \pm 1.6	63 \pm 7
1.75	55 \pm 3	1.88	17.0 \pm 2.3	60 \pm 9
2.0	49 \pm 4	1.82	16.1 \pm 2.2	53 \pm 8
2.5	46 \pm 4	1.80	15.2 \pm 1.7	49 \pm 6
2.75	44 \pm 3	1.78	14.9 \pm 2.0	47 \pm 7
3.0	43 \pm 3	1.77	14.0 \pm 2.3	44 \pm 8
4.0	40 \pm 3	1.75	13.4 \pm 1.5	41 \pm 5

Table 3. Theoretical and experimental values of the coefficient \underline{K}
 $(\text{J/m}^2 / (\text{molecular weight})^{1/2})$ in equation 1

Elastomer	ρ (kg/m^3)	$U \times 10^{19}$ (a) (J)	$q^{1/2}$	l (nm)	M_o (g/g-mole)	\underline{K} (calc. from equation 3)	\underline{K} (expt.)
PB	910	5.75	1.63 (b)	0.115	13.5	0.73	1.15
<u>trans</u> -PI	940	5.75	1.84 (b)	0.127	17	0.67	0.78
<u>cis</u> -PI	920	5.75	1.32 (b)	0.115	17	0.43	1.05
PDMS	970	6.10	2.5 (c)	0.143	37	0.31	0.35
PNF	1,700	3.50	<u>ca</u> 5 (c)	0.160	185	0.07	0.22

^a From R. T. Sanderson, "Chemical Bonds and Bond Energy,"
 Academic Press, Inc., New York, 1971

^b From reference 4.

^c Estimated values.

FIGURE LEGENDS

1. Method of measuring tear strength.
2. Tear strength \underline{T} of natural rubber samples crosslinked with various amounts of dicumyl peroxide and swollen with paraffin oil.
3. Threshold tear strength \underline{T}_0 of various elastomers vs Young's modulus \underline{E} . 1, PB (Δ); 2, cis-PI (\ominus) and NR (O); 3, trans-PI (\bullet); 4, PDMS (\square); 5, PNF (\blacksquare).
4. Threshold tear strength \underline{T}_0 of various elastomers vs molecular weight \underline{M}_c of network strands calculated from Young's modulus \underline{E} by means of equation 4. Symbols as for Figure 3: 1, PB (Δ); 2, cis-PI (\ominus) and NR (O); 3, trans-PI (\bullet); 4, PDMS (\square); 5, PNF (\blacksquare).

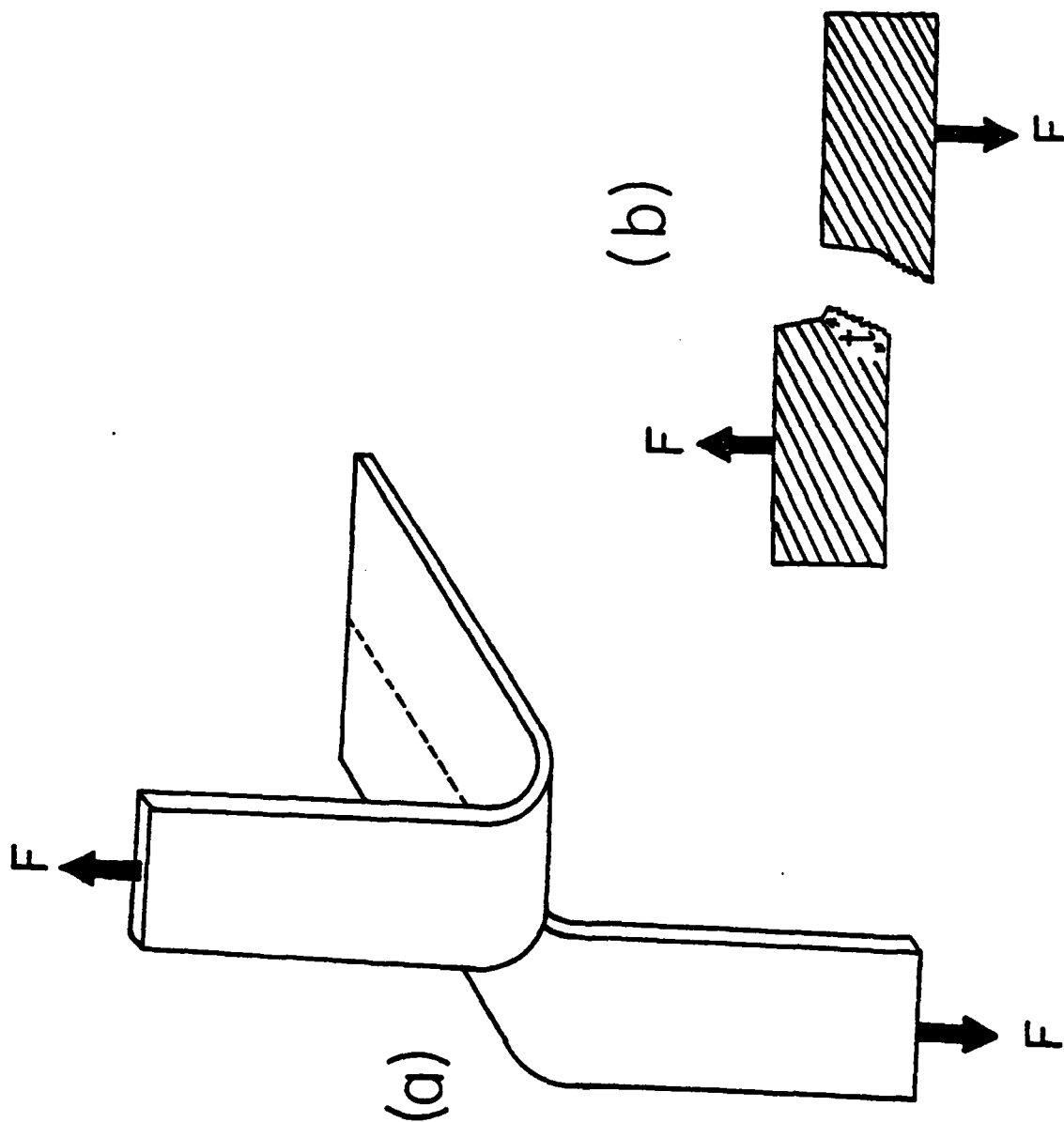


Figure 1.

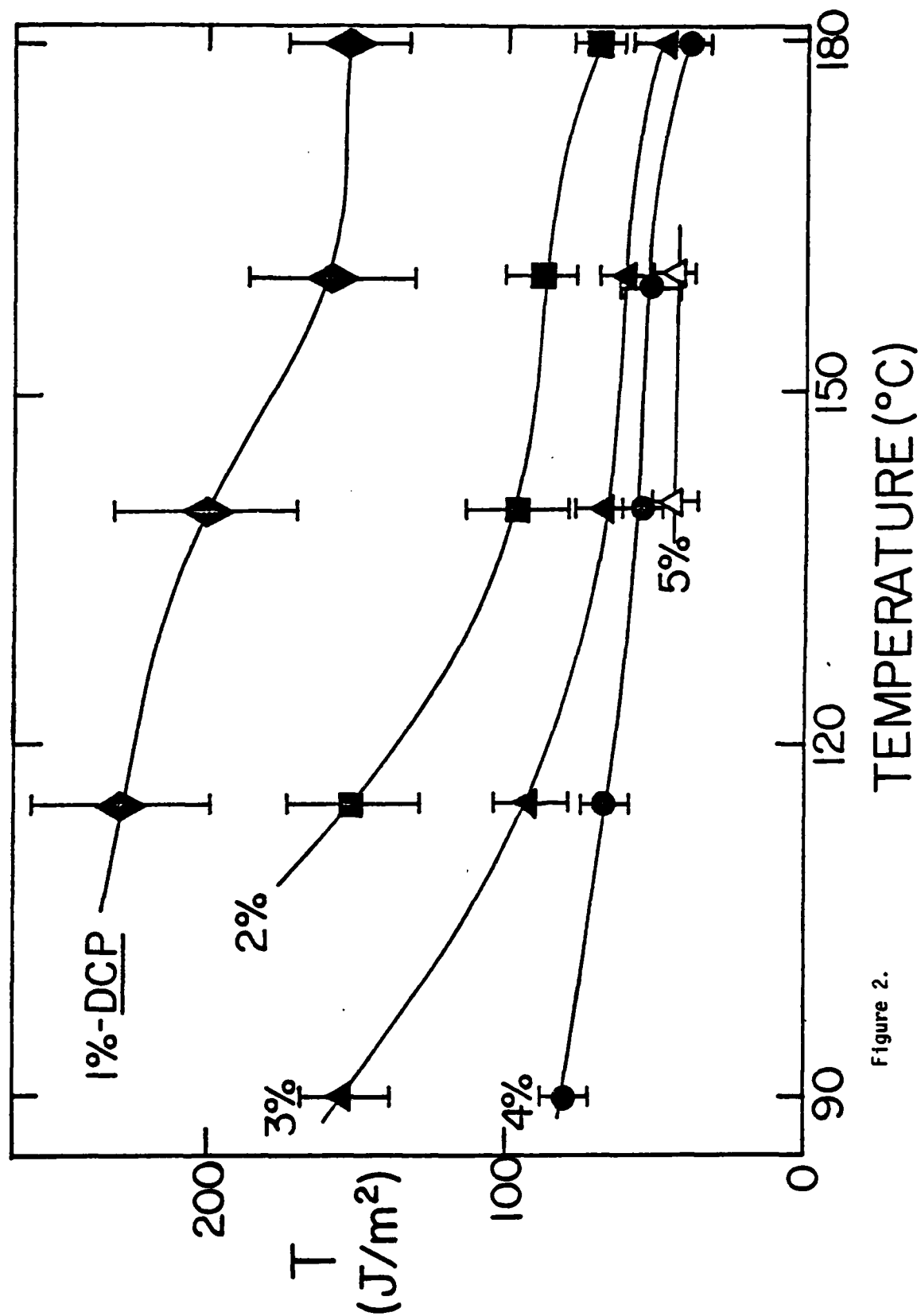


Figure 2.

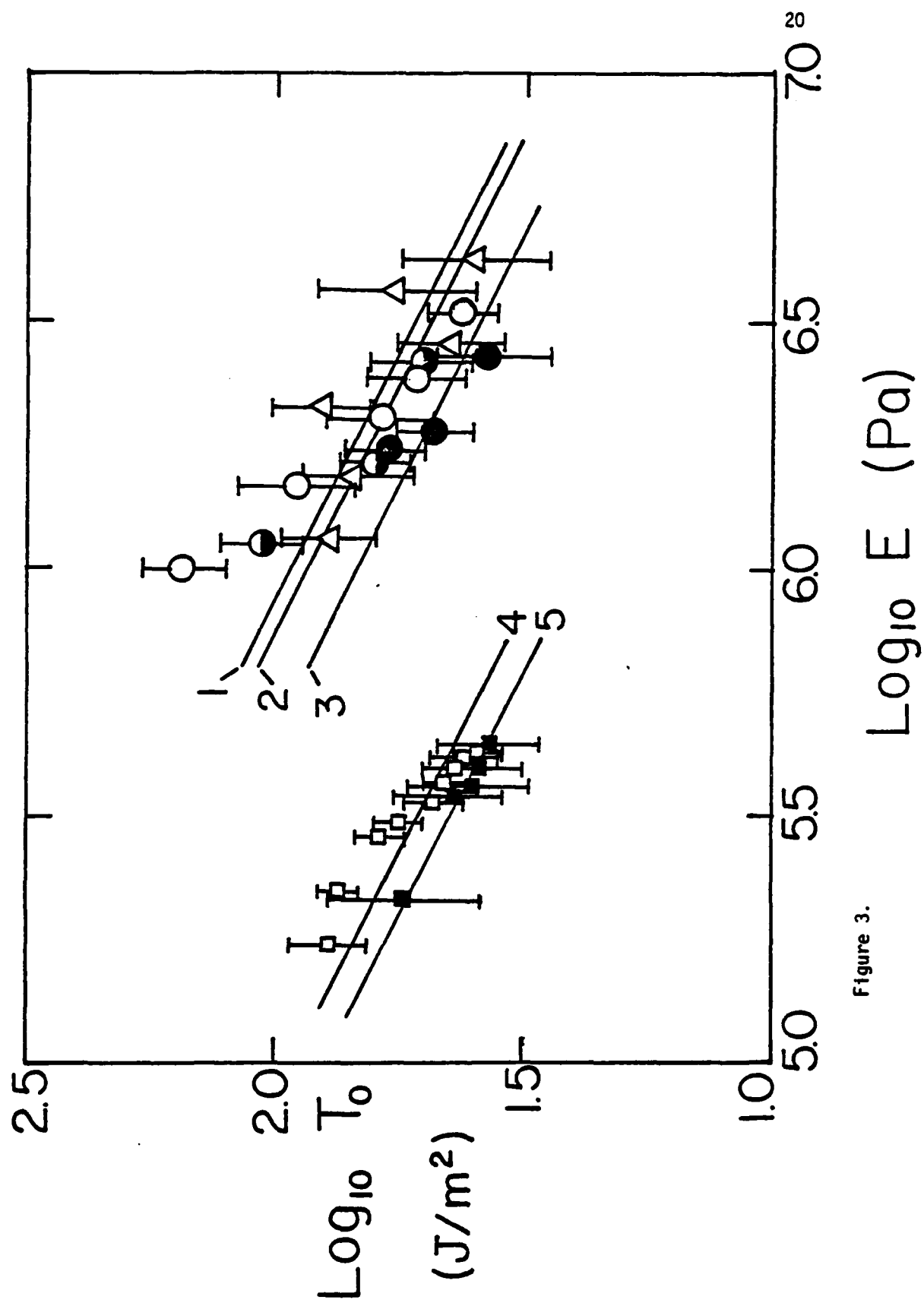


Figure 3.

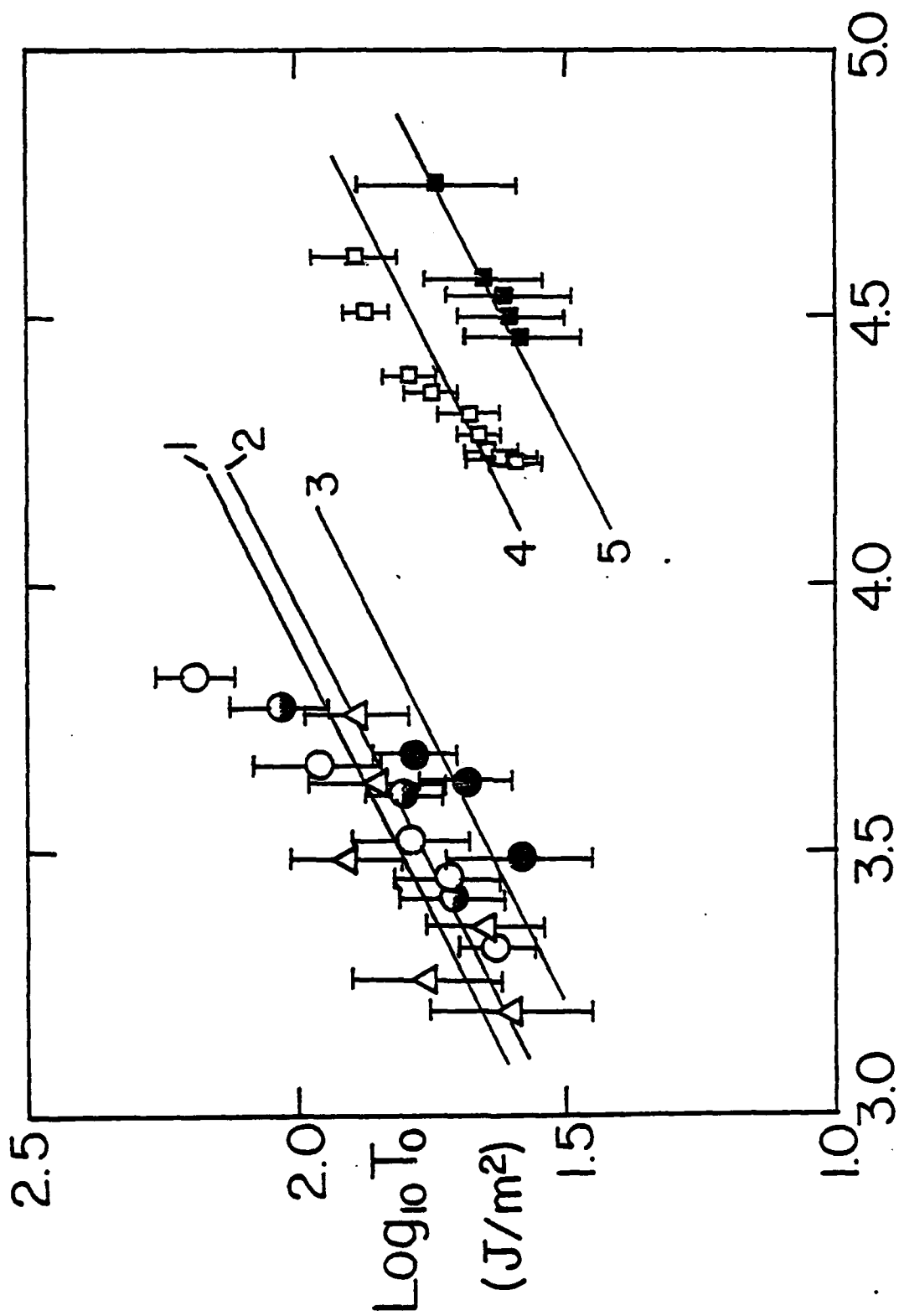


Figure 4. $\text{Log}_{10} M_c$ (g/g-mole)

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